



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

Docket No: 0261-0008

Tou1, et al.

Serial No.: 09/851,983

Art Unit: 1712

Filed : May 10, 2001

Examiner: ZIMMER, MARC S.

Title : TOP COATING COMPOSITION

DECLARATION UNDER RULE 132

Honorable Commissioner of Patents and Trademarks,

P.O. Box 1450

Alexandria, VA 22313-1450

Sir:

I, Hisaki Tanabe, a citizen of Japan and having postal mailing address of 614-8052, Yawata Musashishiba 7-9, Yawata-shi, Kyoto JAPAN, declare and say that:

In March 1977, I was graduated from Faculty of Engineering Science in Osaka University;

I am one of the inventors of the above-identified application and am familiar with the subject matter thereof;

I have read the Official Action mailed and the references cited therein and I am familiar with the subject matter thereof;

I respectfully submit herewith my exact report thereon;

The following experiments were conducted by me or under my direct supervision.

## Experiments

### Synthesis example 1

#### Synthesis of Silicate-grafted resin (I) of the present invention

A one-liter reaction vessel equipped with a thermometer, stirrer, condenser and nitrogen inlet tube was purged with nitrogen gas and then charged with 60 weight parts of MKC Silicate MS-56 (product of Mitsubishi Chemical; average molecular weight 1,500, 22 methoxy groups per molecule) which is a silicate compound, 400 weight parts of hydrolyzable silyl-containing resin D disclosed in the present invention, and 5 weight parts of trimethyl orthoacetate, and the grafting reaction was carried out at 110 °C for 7 hours to give a transparent varnish having a nonvolatile matter content of 65% and containing a silicate-grafted resin (I) resulting from graft polymerization of the silicate compound.

### Synthesis example 2

#### Synthesis of Reacted resin with colloidal silica particle (II) of the reference

A one-liter reaction vessel equipped with a thermometer, stirrer, condenser and nitrogen inlet tube was purged with nitrogen gas and then charged with 200 weight parts of SNOWTEX MIBK-ST (product of Nissan Chemical Industries Co., Ltd., colloidal silica having a particle diameter of 10 to 20 nm and a nonvolatile matter content of 30%, solvent is methyl isobutyl ketone), 400

weight parts of hydrolyzable silyl-containing resin D disclosed in the present invention, and 5 weight parts of 40% aqueous solution of orthophosphoric acid, and the grafting reaction was carried out at 110 °C for 7 hours to give a little turbid varnish having a nonvolatile matter content of 49% and containing a reacted resin with colloidal silica particle (II). By the way, this muddy did not range after 3 hours of reaction and the reaction did not proceed to the particular extent, so the reaction was stopped.

### Synthesis example 3

Synthesis of Reacted product of alkoxysilane compound with colloidal silica particle (III) of the reference

A one-liter reaction vessel equipped with a thermometer, stirrer, condenser and nitrogen inlet tube was purged with nitrogen gas and then charged with 400 weight parts of SNOWTEX MIBK-ST (product of Nissan Chemical Industries Co., Ltd., colloidal silica having a particle diameter of 10 to 20 nm and a nonvolatile matter content of 30%, solvent is methyl isobutyl ketone), 40 weight parts of methyl trimethoxysilane (product of Nippon Unicar Co., Ltd., product name: A-163), and 5 weight parts of 40% aqueous solution of orthophosphoric acid, and the reaction was carried out at 80 °C for 3 hours to give a little turbid solution having a nonvolatile matter content of 35% and containing a reacted product of alkoxysilane compound with colloidal silica particle (III).

### Experiments 1 to 3

A clear coating was prepared by incorporating, in the resin component specified in the following Table 1, 2 weight parts of Tinuvin 900 (ultraviolet absorber, product of Ciba Specialty Chemicals), 1 weight part of Sanol LS-440 (product of Sankyo Company, Ltd.), and 0.1 weight part of Modaflow (surface modifier, product of Monsanto) were added with stirring using a disperser. The clear coating obtained was then diluted with Nippe 500 Standard Thinner (product of Nippon Paint) to a viscosity of 23 seconds (Ford cup No. 4, 20 °C).

A Nippon Paint cationic electrodeposition coating, PowerTop V-20 (trademark), and a polyester-melamine type gray intermediate coating, Orga TO H-870 (each product name), were applied to phosphated steel sheets to dry film thicknesses of 20  $\mu\text{m}$  and 30  $\mu\text{m}$ , respectively and then cured by heating. The coatings were polished with a #2400 sandpaper and cleaned by wiping using petroleum benzine. The thus-prepared coated steel sheets were used as test substrates.

Then, the above diluted clear coating was applied to the substrates to a dry film thickness of 30  $\mu\text{m}$  by spraying at a temperature of 20 °C and a humidity of 70%, followed by 40 minutes of baking and drying at 60 °C to give clear coating films. The coating films obtained were evaluated for appearance and typical performance characteristics. The results are shown in Table 1.

Table 1

	Ex. 1	Ex. 2	Ex. 3
resin component	Silicate-grafted resin (I) of the present invention	56	
	Reacted resin with colloidal silica particle (II)	56	
	Reacted product of alkoxysilane compound with colloidal silica particle (III)		30
	Organic film-forming component	29	52
	OH-containing acryl #1 Polyisocyanate #2	35	48
Amount of silicate and colloidal silica (relative to resin 100)			
Performance	Coating film appearance	20	30
	70% humidity		
Outdoor exposure	Water resistance	O	Turbid
	Initial	O	x Whitened
	Angle of contact with water	76	83
	1 month	-0.2	-3.1
	Angle of contact with water	35	75
	6 months	-0.3	-6.5
	Angle of contact with water	31	65
	Stain resistance of polished part	-0.8	-8.5
			-8.1

\*1: Mitsubishi Rayon's Dianal LR 2562 (OHV = 166, number average

molecular weight = 4,000, nonvolatile matter = 61%)

\*2: Mitsubishi Chemical's Mitec NY 215A (nonvolatile matter = 75%)

※ The above-mentioned evaluation tests were conducted by the same method as disclosed in the present invention.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Signed this 20th day of FEB., 2004

Kisaki Tanaka